Statistically Designed Survey of Polychlorinated Dibenzo-p-Dioxins, Polychlorinated Dibenzofurans, and Co-Planar Polychlorinated Biphenyls in U. S. Meat and Poultry, 2002—2003: Results, Trends, and Implications

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To obtain information on dioxin levels in the human diet, the Food Safety and Inspection Service of the United States Department of Agriculture recently determined levels of dioxin-like compounds (dioxins/dibenzofurans/PCBs) in four major slaughter classes (steers and heifers, market hogs, young chickens, and young turkeys) that comprise over 90% of the meat and poultry production in the United States. The data were analyzed and compared to data from smaller surveys carried out from 1994 to 1996. These surveys were conducted by different laboratories nearly 10 years apart, so a direct comparison of the data was not straightforward. Three approaches were taken: (1) comparison with nondetects set to zero, (2) comparison with nondetects set to half the limit of detection, and (3) comparison applying the earlier surveys' limits of detection to the newer data. The data analyses indicated that dioxin levels appear to have declined in three of the four slaughter classes, with young chickens, market hogs, and young turkeys declining 20-80%, while any declines in cattle dioxin levels, if real, are less than those observed in the other slaughter classes. Further study is needed to examine factors that might explain the differences in dioxin levels and distribution profiles in the four slaughter classes. A small number of market hog and steers/ heifers samples had dioxin toxic equivalency levels (TEQs) greater than 2 pg/g lipid weight. Follow-up investigations

for those samples indicated a common source for the market hog samples (a dioxin-contaminated mineral supplement), but no commonality was found for the steers/heifers samples.

Introduction

In the mid-1990s, the U.S. Department of Agriculture (USDA) and the U.S. Environmental Protection Agency (EPA) conducted surveys of dioxins in beef (1), pork (2), and poultry (3) from slaughter facilities across the United States. The surveys found low levels of dioxin in samples from approximately 50 steers/heifers, 50 market hogs, 41 young chickens, 15 young turkeys, and a small number of samples from minor marketing classes. A survey of 510 beef, pork, and poultry samples was conducted in 2002-2003, in coordination with the U.S. Food and Drug Administration (FDA), EPA, and the USDA's Agricultural Research Service (ARS) to obtain statistically valid information about current levels of dioxins in domestically produced meat and poultry; to investigate any unusual findings to identify possible sources of dioxin into the food supply and to facilitate discussion regarding what steps might be taken to interdict or remove these sources; and to compare results with those from the previous surveys. The new survey of dioxin-like compounds (DLCs) included polychlorinated dibenzo-pdioxins and dibenzofurans (PCDD/Fs) and nonortho polychlorinated biphenyls (PCBs)

The USDA's Food Safety and Inspection Service (FSIS) considers conducting periodic dioxin surveys a prudent public health practice, consistent with the suggestion of the National Academies of Sciences (NAS) study entitled "Dioxins and Dioxin-like Compounds in the Food Supply: Strategies to Decrease Exposure" (4). Such surveys of the food supply can provide insight into changes in environmental levels and human exposure to dioxin-like compounds through dietary components. Surveys can also uncover previously undetected sources of food chain contamination. For example, in the mid-1990s surveys, high dioxin levels in two young chicken samples were traced to ball clay, an anticaking agent used in animal feed (5). The ball clay taken from an open pit mine at a depth of almost 100 feet below the earth's surface was found to be contaminated with a unique pattern of dioxins that suggested natural formation of these dioxins in clay (6, 7) deposited along the shores of the Mississippi Embayment during the early to middle Eocene Epoch, approximately 40-45 million years ago. FDA has since banned the use of ball clay in feed.

DLCs enter the environment as byproducts of combustion and manufacturing processes such as power plants, paper manufacturing, and municipal and medical waste incineration. DLCs are persistent and remain in the environment for decades (8). They accumulate in the fatty tissues of humans and animals. Over 90% of human dioxin exposure is a result of dietary intake of animal fats and fish (9). Based on strong evidence for cancer in animals, 2,3,7,8-tetrachlorodibenzo-p-dioxin is labeled a "known human carcinogen" (10), and other DLCs are widely thought to be "likely carcinogens" (8). The controversy concerning the strength of the evidence for health effects in humans continues, although data indicating carcinogenicity and possible health effects to human immune and endocrine systems, as well as fetal and child development, are accumulating (11–13).

Since the 1994-1996 surveys, EPA implemented a number of policies and actions aimed at lowering dioxin emission

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TABLE 1. Toxic Equivalency Factors (TEFs) and Average Concentrations of Seventeen PCDD/Fs, Three Co-Planar PCBs, and TEQs in Each Slaughter Class^a

congener	TEF	steers/heifers n = 139	market hogs $n=136$	young chickens $n = 151$	young turkeys $n = 84$
2378-TCDD	1	0.06 (0.04)	0.04 (0.00)	0.04 (0.01)	0.06 (0.03)
12378-PeCDD	1	0.23 (0.23)	0.03 (0.02)	0.06 (0.05)	0.17 (0.17)
123478-HxCDD	0.1	0.30 (0.30)	0.08 (0.07)	0.05 (0.04)	0.10 (0.10)
123678-HxCDD	0.1	1.63 (1.63)	0.18 (0.17)	0.26 (0.25)	0.37 (0.37)
123789-HxCDD	0.1	0.32 (0.32)	0.03 (0.00)	0.06 (0.04)	0.05 (0.03)
1234678-HpCDD	0.01	3.97 (3.97)	1.20 (1.19)	1.23 (1.22)	0.23 (0.20)
OCDD	0.0001	3.92 (3.24)	9.14 (8.57)	4.97 (4.36)	2.18 (1.32)
2378-TCDF	0.1	0.03 (0.01)	0.02 (0.00)	0.07 (0.06)	0.17 (0.17)
12378-PeCDF	0.05	0.05 (0.00)	0.05 (0.00)	0.07 (0.03)	0.09 (0.06)
23478-PeCDF	0.5	0.16 (0.15)	0.08 (0.07)	0.06 (0.06)	0.16 (0.16)
123478-HxCDF	0.1	0.41 (0.40)	0.17 (0.14)	0.09 (0.06)	0.09 (0.08)
123678-HxCDF	0.1	0.25 (0.23)	0.13 (0.08)	0.08 (0.04)	0.08 (0.04)
234678-HxCDF	0.1	0.21 (0.19)	0.09 (0.05)	0.06 (0.02)	0.05 (0.01)
123789-HxCDF	0.1	0.03 (0.00)	0.03 (0.00)	0.03 (0.00)	0.02 (0.00)
1234678-HpCDF	0.01	0.81 (0.75)	0.68 (0.60)	0.21 (0.13)	0.12 (0.03)
1234789-HpCDF	0.01	0.05 (0.04)	0.05 (0.04)	0.02 (0.01)	0.02 (0.00)
OCDF	0.0001	0.15 (0.11)	0.44 (0.29)	0.15 (0.11)	0.11 (0.07)
PCB-77	0.0001	3.59 (0.89)	3.58 (0.64)	5.19 (2.91)	3.73 (1.26)
PCB-126	0.1	1.23 (1.23)	0.20 (0.18)	0.68 (0.68)	1.69 (1.69)
PCB-169	0.01	0.32 (0.32)	0.30 (0.27)	0.38 (0.36)	0.79 (0.78)
TEQ D/F (ppt)		0.74 (0.70)	0.21 (0.13)	0.22 (0.15)	0.41 (0.37)
TEQ PCB (ppt)		0.13 (0.13)	0.02 (0.02)	0.07 (0.07)	0.18 (0.18)
total TEQ (ppt)		0.87 (0.83)	0.23 (0.15)	0.29 (0.22)	0.59 (0.55)
TEQ range (ppt)		0.17-6.08	0.09-4.41	0.09-1.87	0.13-1.81
O = 4FF -7		(0.08 - 6.08)	(0.00 - 4.41)	(0.00-1.82)	(0.04 - 1.80)
		•	•	•	•

^a Levels are reported in pg/g lipid with nondetects = 0.5 × LOD and nondetects = 0 in parentheses.

levels from incinerators and other stationary sources, although dioxins continue to recycle through the environment (I). Because the 1994-1996 data were outdated and of limited use and applicability to today's environmental situation, a new, larger survey of the four major categories of meat and poultry consumption was undertaken to provide a more useful basis for future policy considerations.

In June 2005, FSIS posted the initial results from the 2002—2003 survey on its web site (14). These results were not background subtracted. Because the previous USDA-EPA surveys presented background-subtracted data, this manuscript also presents background-subtracted data permitting a more thorough comparison of the data from the surveys.

Experimental Section

Steers/heifers, market hogs, and poultry were included in this survey because these groups represent slightly more than 90% of the meat and poultry produced in the United States. Since 90% of this meat and poultry is consumed domestically, production was used as a surrogate for consumption.

A total of 510 samples were collected and analyzed from slaughter establishments across the continental U.S.: 139 steers/heifers, 136 market hogs (gilts and barrows), 151 young chickens, and 84 young turkeys

All establishments actively slaughtering animals in the four production classes were included in the sampling frame. The number of samples collected from each facility was proportional to the plant's production volume for that slaughter class in the preceding 12-month period. Thus, large slaughter establishments had a greater probability of being sampled than smaller ones; randomly selected samples within a class should be representative of the entire class. The sampling frame was updated quarterly during the survey, with sampling beginning May 2002 and ending May 2003.

The total number of samples per slaughter class had to provide a sufficient number of samples, within the resource constraints to ensure that the results would be representative of the levels found within the U.S. meat and poultry supply. Thus, young turkeys and market hogs were over-sampled relative to their proportion of the meat and poultry supply, while young chickens were slightly under-sampled.

FSIS inspectors collected approximately 250 g samples of back fat from steers/heifers, belly fat from market hogs, or abdominal fat from the young chickens and young turkeys. Poultry samples were composites from three birds in the same flock. Samples were frozen, shipped in sealed boxes to the USDA ARS Biosciences Research Laboratory, Fargo, ND, and stored at $-60~^{\circ}\mathrm{C}$ until the time of analysis.

PCDD/Fs and nonortho PCBs were analyzed by a modification of EPA Method 1613B (tetra- through octa-chlorinated dioxins and furans by isotope dilution HRGC/HRMS) previously described for trimmed fat (15). A blank and a known spiked sample were analyzed with every eight survey samples to provide ongoing quality assurance for the method. Because some congeners were present in low but detectable amounts in the method blanks, the blank levels were subtracted from each sample in the corresponding set.

Toxic equivalency values (TEQs) were calculated from the data using the 1998 World Health Organization (WHO) toxic equivalency factors (TEFs) (16), and setting nondetects equal to zero or one-half the limit of detection. The limit of detection (LOD) was determined at the 95% confidence level as 2 times the standard deviation of the blanks or of a replicate low-level spike if a congener was not routinely present in the blanks by the method described by Glaser et al. (17). The limit of quantitation (LOQ) was determined in a similar manner using 10 times the standard deviation.

Results and Discussion

Dioxin Levels in Different Slaughter Classes. All references to results from cattle, hogs, chickens, and turkeys in the 2002—2003 survey refer to results from steers/heifers, market hogs, young chickens, and young turkeys. Table 1 summarizes the analytical data by slaughter class and provides the average concentration for each congener and the slaughter class average TEQ calculated when nondetects (nd) = 0 or 0.5 \times

LOD. Cattle had the highest average levels of PCDD/F TEQ and total TEQ (PCDD/F and PCBs) of any slaughter class, followed by turkeys. Chickens and hogs had the lowest average TEQ levels for dioxins, furans, and PCBs. At first glance, the higher average dioxin level observed in cattle might be explained solely by age. They reach a marketable weight in approximately 18–24 months, a longer period of time than for the other slaughter classes, and therefore, potentially have more time to accumulate dioxins from the environment and diet.

However, hogs, which are slaughtered at approximately 6 months of age, have virtually the same average TEQ for DLCs as chickens, which are slaughtered at about 1.5 months (Table 1). Additionally, hogs have only half the average dioxin TEQ of turkeys, which are slaughtered when they are 4–5 months old. Hogs have slightly lower PCB levels than chickens, while turkeys have the highest level of PCBs of the four slaughter classes. When viewed from the perspective of TEQ accumulation/month chickens (1.5 months) and turkeys (4–5 months) have the highest relative levels of dioxins.

Examining alternative scenarios, such as dietary composition and production environment, feed source, intake and feed efficiency, and percent carcass fat, provides insight into the relative levels of DLCs found in the four slaughter classes.

Dietary Composition and Production Environment. Aerosol deposition is known to lead to DLC contamination on the surfaces of forages providing an entrance into the food supply (18, 19). Grains such as soybeans and corn should not be subject to contamination from aerosol deposition because outer covers (seed pods, husks) are removed prior to feeding. Cattle have a high percentage of forages in their diets, whereas poultry and hogs have largely grain-based diets. Prior to being placed in feedlots for finishing, cattle also graze on grasses. It has been reported that grazing cattle can ingest up to several kg/day of soil (20), which may be contaminated by dioxins from past deposition. In contrast, the indoor production environment for most poultry and hogs minimizes their contact with soils.

The differences between a diet of forages for cattle, including possible soil ingestion, versus a grain-based diet for hogs and poultry, could provide part of an explanation for the observed differences in DLC levels. This factor (forage vs grain), however, does not explain why turkeys have an average TEQ approximately twice that found for chickens and hogs.

Turkey diets, however, contain significantly higher levels of fat than that of chicken, cattle and hogs—and that fat can come from recycled sources. The NAS Committee viewed fats as a likely recycling factor for dioxins within the food chain because of the solubility of these substances in fat (4). The presence of higher levels of fat in the diet of turkeys could increase dioxin levels in two ways: the higher fat content could serve as a larger potential source for DLCs than grain or forages, and the presence of higher levels of lipids in the diet could facilitate the absorption of these fat soluble substances from other sources, as suggested in rodent dosing studies (21).

Feed Source, Feed Intake, and Feed Efficiency. Dietary regimens, which include feed source and the inter-relationship between feed intake and feed efficiency (efficiency of weight gain), could play a role in average slaughter class dioxin levels. The role that a roughage-based diet vs a grainbased diet plays has been presented above, but all animal diets also contain animal or aquatic fats (fish meal). Dioxin levels in fish meals vary widely, and the role the geographic source of that fish meal plays has been reported previously (22).

Different market weight requirements also impact the amount of feed an animal requires to reach its market weight.

Additionally, differing feed efficiencies will mean that the different slaughter classes will require differing amounts of feed to gain one pound in body weight. For example, cattle consume approximately 6–10 pounds of feed (dry weight) for each pound of carcass weight gain, whereas poultry and swine are more feed efficient, consuming less feed per pound of carcass weight gain (23–25). Cattle will, therefore, consume more feed with the potential to sequester more dioxins in their body fat by the time they reach market weight. The lower feed intake/feed efficiency factors for cattle could explain why they have the highest absolute dioxin levels of the four slaughter classes.

Size of Fat Pool/Percent Body Fat. There is a wide variation in percent body fat among the four slaughter classes, ranging from 7.8% (turkeys) to 30% (market hogs). Although large amounts of body fat may be considered a diluent for dioxins in the body, this conjecture is not supported by bioconcentration studies in dairy cows, beef cattle, chickens, and hogs (26–29). These different slaughter classes bioconcentrate dioxins from the diet into lipid stores to the same degree, despite the wide range of percent body fat in these classes. Similar bioconcentration factors imply that similar dioxin levels in the feed will produce similar levels in the adipose tissue of each slaughter class. The differences in dioxin levels observed in this study are probably not related to percent body fat but to ingestion.

Congener Profiles. In general, average congener concentrations were in the low ppt range (Table 1). Turkeys and chickens appear to have higher levels of TCDF (0.17 and 0.07 ppt respectively) than cattle and hogs (0.03 and 0.02, resepectively, for TCDF). In addition, TCDF was detected in over 60% of the poultry samples but in less than 10% of either the cattle or hog samples. Chickens and turkeys also had a somewhat greater contribution of coplanar PCBs to their total TEQ, i.e., 24% and 30%, than did cattle (15%) or hogs (9%). This may reflect lower metabolizing capabilities in poultry for these compounds (30), or differences in dietary inputs (fishmeal, minerals, etc.) or production practices.

One of the congeners with the highest individual toxicity, 2,3,7,8-TCDD, was not detected in 87% of samples and contributed only 7-17% to the total TEQ when nondetects were set equal to $0.5 \times \text{LOD}$. Three congeners (1,2,3,7,8-PeCDD,2,3,4,7,8-PeCDF, and PCB-126) contributed 40-70% to the total TEQ in each animal class whether nondetects were set equal to zero or $0.5 \times \text{LOD}$. Each of these three congeners represented 9-29% of the total TEQ. Another congener that contributed significantly to the cattle total TEQ was 1,2,3,6,7,8-HxCDD (19%).

1,2,3,7,8-PeCDD and 2,3,4,7,8-PeCDF dominate the TEQ partly because they have the highest toxicity factors (TEF = 1.0 and 0.5, respectively). In addition, these congeners have higher bioavailability than hexa, hepta, or octa congeners; are not as readily metabolized as TCDF and 1,2,3,7,8-PeCDF; and are found in many anthropogenic sources (21, 31-33). 1,2,3,6,7,8-HxCDD was the most prevalent hexa-congener found in the samples and accounted for 6-19% of the TEQ. This congener was previously observed to be the dominant hexa-congener in foods (34, 35). As a contaminant found in the wood preservative pentachlorophenol, it has been implicated as a source of dioxin exposure in cattle (36, 37) due to its relatively high bioavailability compared to other dioxins found in pentachlorophenol (18% vs 3% for HpCDD and 0.4% for OCDD) (31).

TEQ Distribution within Slaughter Classes. The histograms in Figure 1 show the distribution of the individual animals in each slaughter class over the TEQ range. Most of the hog and chicken samples cluster near the lower end of the TEQ range with only a few animals over 1.0 ppt. These distributions suggest fairly homogeneous population exposures. The findings for these two slaughter classes are not

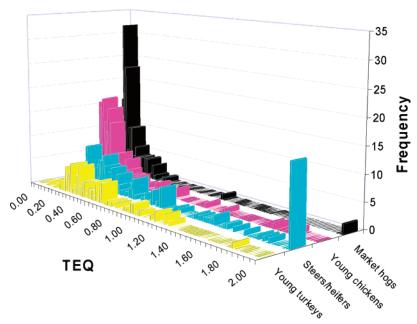


FIGURE 1. Distribution of individual samples in each class over the range of total TEQs (pg/g lipid).

inconsistent with confinement production practices which are expected to reduce exposure to soils and other non-controllable (environment-related) variables. Additional factors could include similar rearing conditions and, in the case of young chickens, a short growth period (approximately 6 weeks) that would minimize the chance of an intervening contamination event leading to a broader range of TEQ distributions.

Cattle and turkey results were distributed over a broader TEQ range. For cattle this may not be surprising as they are raised on a larger variety of grazing areas and feedlot conditions; they are far more likely to be transported from one area of the country to another for finishing in feed lots than the other three slaughter classes. For turkeys, however, the lack of a homogeneous distribution of results was not expected. Although much of the turkey production industry has moved toward confinement production in the past decade, turkeys are still more likely to have greater contact with soil than chickens or hogs. Soil ingestion plus the higher percentage of animal or fish fat in the diets of turkeys vs chickens or hogs are possible explanations for their broader TEQ distribution range.

Close examination of the distributions shows that the data are not normally distributed (Figure 1). Several statistical (nonparametric) tests were performed to determine whether differences between the PCDD/F and PCB TEQ distributions of the different slaughter classes are statistically significant. The results from the Kolmogorov–Smirnov (K–S) test, which examines uniqueness of distributions, showed several highly significant results (p-values <0.0001), indicating the improbability of obtaining such test values simply by chance if there were no true difference in distributions. Thus, most of the cross-slaughter class distributions are independent of one another (cattle-hog, cattle-chicken, chicken-turkey, and hog-turkey). Several paired comparisons however, have higher p-values (0.28–0.45), indicating greater likelihood that the distributions are similar (PCDD/Fs for chicken-hog, barrow-gilt, and steer-heifer). The cattle-turkey PCDD/F TEO distributions are of intermediate independence, p-value = 0.008.

A second test, the Wilcoxon rank-sum test, provides overall results similar to those of the K-S test, but suggests a greater degree of similarity between the cattle and turkey distributions, while the chicken-hog distribution showed a lesser

degree of similarity. The two tests showed different p-values for the steer—heifer PCDD/F TEQ distributions (K-S p-value = 0.28; Wilcoxon p-value = 0.08).

A small difference in average total TEQ values for steers and heifers was observed (steers, 0.78 ppt; heifers, 1.00 ppt). The difference, bordering on the accuracy of the method for most congener analyses (\pm 25%), might be real because the comparison is between averages, not individual analyses. The difference could also be related to sample sizes; a larger number of steers (83) were sampled than heifers (56). Similar differences were not noted in the sampled hog population (72 barrows, 64 gilts), in which the numbers of barrows and gilts sampled were more evenly balanced. Barrows and gilts averaged 0.188 and 0.191 ppt TEQ, respectively. The apparent difference between steers and heifers lies within the dioxin/ furan TEQ contribution rather than the PCB TEQ. Reasons for the difference are unclear, but future studies could investigate whether this apparent gender difference in cattle is real or not.

Comparison of New Data with Previous U.S. Surveys. A major objective of this survey was to compare the new results with the results of the surveys conducted in the mid-1990s and determine if the levels of PCDD/Fs and coplanar-PCBs have changed. To attempt a comparison, data from the 2002-2003 survey were blank-subtracted as done in the earlier surveys; however, differences still exist between the surveys such as laboratory environments, instrument capabilities, cleanup methods, and decision criteria, making an exact comparison difficult. The main complicating factor is related to the limits of detection (LODs) in the surveys. The LODs in the old surveys were determined differently from those in the new survey, leading to higher LODs for many congeners (38); one congener LOD was approximately 20 times higher in the earlier surveys than in the 2002-2003 survey. Higher LODs produce lower TEQ values when nondetects are set to zero and higher TEQ values when nondetects are based on the LOD

To examine the impact these LOD differences have on the results, we compared the data using three different approaches: (1) with nondetects set equal to zero, (2) with nondetects set equal to 0.5 \times LOD, and (3) by applying the older LODs to the newer data set. The data in Table 2 show the results for the first two comparisons. Hogs, chickens, and turkeys had similar percentages of nondetected con-

TABLE 2. Comparison of Surveys of Meat and Poultry from Mid-1990s and 2002—2003

class	mid-1990s surveys			2002-2003 survey			change in TEQ	
	TEQ nd = 0	$\begin{array}{c} \text{TEQ} \\ \text{nd} = \text{0.5} \times \text{LOD} \end{array}$	% nd	TEQ nd = 0	$\begin{array}{c} \text{TEQ} \\ \text{nd} = \text{0.5} \times \text{LOD} \end{array}$	% nd	nd = 0	$nd = 0.5 \times LOD$
young chickens	0.65	0.94	55%	0.22	0.29	63%	-66%	-69%
young turkeys	1.32	1.53	51%	0.55	0.59	52%	-58%	-61%
market hogs	0.40	1.47	82%	0.15	0.23	72%	-63%	-84%
steers/heifers	0.68	1.38	78%	0.83	0.87	44%	+22%	-37%

 $^{^{}a}$ Average TEQ values in pg/g lipid for dioxins, furans, and PCBs when nondetects (nd) = 0 or 0.5 \times LOD. The percent of nondetected congeners in each survey is given along with the change in total TEQ.

geners in all surveys, and the average TEQs for these three slaughter classes are approximately 60-80% lower in the new survey than in the older ones, whether based on nd = 0 or nd = $0.5 \times \text{LOD}$. This finding strongly suggests that dioxin levels for these three slaughter classes have, in fact, decreased during the past decade.

For cattle, a comparison of the TEQ values from both surveys with $nd=0.5\times LOD$ shows that the average TEQ level for cattle decreased by 37%. However, when nd=0, the average TEQ increased by 22% in the 2002-2003 survey. Thus, the analysis of cattle data shows that average TEQ values have either decreased or increased, depending on how the nondetect results are treated. The analysis is complicated by both the difference in the LODs and the fact that there were substantially higher numbers of nondetected congeners for steers/heifers in the earlier survey (78%) than in the more recent survey (44%).

The third approach for comparing data from the surveys is to treat the 2002–2003 results as if they had the same LODs as the data obtained in the mid-1990s. When this analysis is performed using nd = $0.5 \times \text{LOD}$, average cattle and hog TEQs show decreases of 12% and 18% respectively, while chickens and turkeys show decreases of 45% and 55% from the mid-1990s survey TEQ values. Making the same calculation when nd = 0 yields TEQ declines of 75–80% for hogs, chickens, and turkeys, while cattle decline by 21%.

Overall, average TEQs over the past decade have decreased for hogs, chickens, and turkeys, regardless of which approach was used to handle the nondetects. The analyses lead to different results for cattle depending on the analytical approach utilized. Even with the two approaches that show declining averages TEQs for cattle, the percentage declines are considerably less for cattle than for hogs, chickens, and turkeys. Based on this "three-approach" analysis, it appears that any decline in the level of dioxins in cattle, if real, is less than that observed in the other slaughter classes.

Examination of the actual distribution of TEQ values for all surveys may also provide additional insight into whether cattle TEQs have actually declined. Both surveys have a similar percentage of steers and heifers with total TEQ levels greater than 2 ppt (nominally the 90th percentile of the mid-1990s surveys): 11% (15 of 139) in the new survey vs 16% (8 of 51) in the old survey. None of the other slaughter classes exhibited such a high percentage of TEQ values at the high end of their distribution curves. In fact, as can be seen in the histograms (Figure 1), the TEQ distribution for steers/heifers in the new survey is quite different from that of the other slaughter classes, particularly when compared to chickens and hogs.

A comparison of congener profiles from the mid-1990s surveys with the current survey indicated few major changes. 1,2,3,7,8-PeCDD, 2,3,4,7,8-PeCDF, and PCB-126 were the dominant congeners in both sets of data, with 1,2,3,6,7,8-HxCDD another significant contributor in cattle (10% of TEQ in mid-1990 and 19% of TEQ in 2002–2003). Because the basic congener profiles for each slaughter class appear to remain reasonably constant since the mid-1990s, the sources

of dioxin exposures may be the same, although the overall levels of these substances in these sources may have decreased.

General Comments. Over 89% of the samples in the new survey (455/510) had total TEQs less than 1.0 ppt, and 97% were less than 2.0 ppt (nd = $0.5 \times \text{LOD}$). No chickens or turkeys exceeded a total TEQ value of 2 ppt TEQ. PCB levels were low—only 6 animals had PCB TEQs greater than 0.5 ppt, and only one of those, a steer, had a PCB TEQ greater than 1.0 ppt. This steer had the highest PCB level and was the only animal in the survey that had both PCDD/F and PCB TEQ levels each greater than 1.0 ppt. Seventeen samples were found with total TEQs between 2 and 6 ppt, including eight steers, seven heifers, and two barrows, and were selected for follow-up investigations. The fifteen cattle with TEQs > 2.0 ppt originated in 10 different states across the country, and there was no discernible connection among these animals.

During four of the follow-up investigations, samples of treated fence or barn posts and wooden feed troughs which showed evidence of being gnawed or rubbed, and bedding materials were obtained. They were found to contain elevated levels of dioxins. The patterns were not inconsistent with PCP-treated wood being the source of the elevated dioxin levels in the cattle, although it is also possible that there could be additional contributing sources to the elevated levels. Although most of the cattle selected for follow-up (TEQ > 2 ppt) were traced back to feedlots, only four could be traced back to the original farms. Several of these animals could only be traced back to an auction barn prior to being sent to slaughter.

The two barrows selected for follow-up (TEQ > 2 ppt) were traced to a common source. Both were raised in Iowa under the same management structure (similar management and husbandry practices, including common feed and feed supplements), although the farms were approximately 100 miles apart. The samples were collected within four weeks of one another and showed similar congener patterns. Further investigation revealed that a dioxin-contaminated mineral supplement was used in the feed and was the likely source of the elevated PCDD/Fs in these two animals. This mineral supplement contamination was also implicated by the Food and Drug Administration as a possible source of contamination in livestock, fish, and poultry (http://www.fda.gov/bbs/ topics/ANSWERS/2003/ANS01203.html) and was removed from the market. No other food animals in the survey indicated both elevated levels and a similar congener pattern to that of the two barrows.

If the new survey data were considered within a broader context such as the European Union's maximum limit paradigm (European Commission Regulation (EC) No 199/2006), only five of 139 cattle exceeded the EU's maximum dioxins and furans limit of 3 ppt TEQ for ruminants; two of them by approximately 10%, a value that is approximately the same as the quantitative reproducibility of the method at this TEQ level. Three of 136 hogs exceeded the EU dioxins

and furans limit of 1 ppt TEQ; two of those animals were identified with a dioxin contamination from a mineral supplement. The third hog exceeded the limit by less than 10%. None of the chickens or turkeys exceeded the EU limit of 2 ppt TEQ. Thus, one could conclude that the U.S. meat and poultry supply compares quite favorably with EU MRLs in terms of average TEQ levels for dioxins and related compounds.

In summary, USDA completed an extensive survey of DLCs in four major slaughter classes. Average dioxin levels appear to have decreased in hogs, chicken, and turkeys, regardless of how nondetects are treated in the analysis. The results for cattle, however, are equivocal, dependent on how nondetects are treated. Several dietary input and environmental factors were discussed to explore how they might impact dioxin levels in the four slaughter classes. A possible gender difference in average dioxin levels was found in the steers/heifers data. Future research focused on these and other factors related to dioxin exposure routes is planned at the UDSA with the goal of reducing dioxin levels in meat and poultry products.

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Supporting Information Available

Eleven supplemental tables providing data on the sampling frame, probability values, method blanks, individual congener LODs and LOQs, and all individual sample TEQs (nd = 0 and nd = 0.5 \times LOD; background-subtracted and nonbackground-subtracted), and a figure comparing congener patterns in the mid-1990 and 2002–2003 surveys. This material is available free of charge via the Internet at http://pubs.acs.org.

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